INEEL/CON-99-00110
PREPRINT

Preliminary Environmental Flow and Transport Modeling at the INEEL

S. O. Magnuson
J. M. McCarthy
J. D. Navratil

September 26, 1999 – September 30, 1999

7th International Conference on Radioactive Waste Management and Environmental Remediation (ICEM '99)

Preliminary Environmental Flow and Transport Modeling at the Idaho National Engineering and Environmental Laboratory, USA

Swen O. Magnuson, James M. McCarthy, and James D. Navratil
Lockheed Martin Idaho Technologies Company
Idaho National Engineering and Environmental Laboratory

ABSTRACT

The Idaho National Engineering and Environmental Laboratory (INEEL) is located in southeastern Idaho in the USA. The primary mission since the laboratory was founded in 1949 has been nuclear reactor research. Fifty-two reactors have been built and operated on the INEEL. Other principal activities at the laboratory have been reprocessing of spent nuclear fuel. Low-level radioactive waste generated on site and mixed and transuranic waste from the Rocky Flats plutonium processing facility in Colorado has been disposed on the INEEL at the Radioactive Waste Management Complex (RWMC). Waste disposal at the RWMC began in 1952 with shallow land burial in pits and trenches. The INEEL was placed on the National Priorities List (NPL) in 1989. The resulting environmental assessments of the potential negative health impacts of disposed waste at the RWMC have required the use of predictive numerical simulations. A petroleum reservoir simulator called TETRAD was modified for use in simulating environmental flow and transport. Use of this code has allowed the complex subsurface stratigraphy to be simulated, including an extensive region of unsaturated fractured basalt. Dual continual simulation approaches have been used to assess combined aqueous- and gaseous-phase transport of volatile organic compounds as well as dissolved-phase transport of radionuclides. Calibration of the simulator to available monitoring data has increased the confidence in the simulator results to the point where the model sensitivities are being used to direct additional characterization efforts. Eventually, as the model calibration improves and confidence in the model predictions increases, the simulator will be used as a decision tool for selecting remedial alternatives for the wastes buried at the RWMC. An overview of the overall program including a summary of laboratory actinide migration studies will be presented.

This work was funded by the U.S. Department of Energy under DOE Idaho Operations Office Contract DE-AC07-94ID13223.

INTRODUCTION

Simulations of flow and transport are used in regulatory environmental assessments conducted at the INEEL. The simulations are necessary to make predictive estimates of future environmental health hazards associated with wastes that have been disposed at selected locations on the INEEL. The complex subsurface lithology and hydrology and geochemical environments has required the development of advanced simulation capabilities. One example of such an application is at a facility called the Radioactive Waste Management Complex (RWMC).

In the remainder of this paper, the background of the RWMC is briefly described along with the development of the conceptual model describing flow and transport at the RWMC. The implementation and calibration of the simulation model is discussed. The use of the sensitivity of the predictive results from the model in directing additional characterization work are summarized.

BACKGROUND

The RWMC is located in the southwest portion of the INEEL in southeastern Idaho in the USA. Low-level, mixed, and transuranic radioactive wastes were buried in shallow pits and trenches in the SDA from 1952 until 1970. Since 1970, transuranic waste has been stored in above-grade facilities. Currently, low-level waste is still being disposed in a portion of the SDA. The SDA overlies a thick stratigraphic sequence composed primarily of fractured basalts that are occasionally interrupted by sedimentary interbeds that were deposited during extended periods of volcanic quiescence. The three primary sedimentary interbeds beneath the SDA occur at depths of approximately 10, 30, and 70 m below land surface. Both the upper surface and thickness of the interbeds vary spatially. There are deeper interbeds beneath the SDA but the limited wellbore information indicates they are much less continuous. The Snake River Plain Aquifer (SRPA) underlies the SDA beginning at a depth of approximately 175 m below land surface.

A simulation study was conducted to estimate fate and transport of contaminants in wastes disposed in the RWMC. The complete results of the simulation study are documented in Magnuson and Sondrup (1998). The overall approach was to develop a simulation model representative of flow and transport in the subsurface. Representativeness was obtained and demonstrated by calibrating model results to observed water and contaminant data and is a substantial improvement over earlier simulation studies.

CONCEPTUAL MODEL

The conceptual model used for the flow simulation consisted of spatially variable and time-dependent infiltration at the surface of the SDA. Infiltrating water moved downward under the influence of gravity and capillarity. Downward movement occurred within both the fractured basalt flows and sedimentary interbeds that compose the vadose zone beneath the SDA. The sedimentary interbeds had both variable thickness and variable elevations to the overlying and underlying basalt flow contacts. Perched or near-perched water conditions occurred in association with the sedimentary interbeds when adequate water was either supplied from above or through horizontal movement within the vadose zone. Water movement within the fractured basalts was considered to be controlled primarily by the fracture network. Water that infiltrated at the surface eventually reached the SRPA. Horizontal movement of water within the SRPA caused the water that originally infiltrated at the SDA surface to move laterally within the aquifer.

The transport conceptual model assumed contaminant movement is controlled primarily by advection and dispersion. Because the flow conceptual model consisted of water movement primarily within the higher permeability fractures and rubble zones in the basalt portions of the subsurface, sorption of contaminants onto surfaces within the basalt matrix was considered not to occur. Rather, sorption occurred onto fine-grained sediments or chemical and weather alteration products coating the surfaces of the fractures through which water and contaminants moved. Sorption onto these fracture coatings was estimated based on assumed effective fracture geometries. Including sorption in this manner implied that the basalt matrix did not effect dissolved-phase transport. This is consistent with the simulation results (Magnuson 1995) from the Large Scale Aquifer Pumping and Infiltration Test conducted near the SDA (Wood and Norrell, 1996). Updated estimates of the sediment sorption properties for each contaminant were used. Diffusion of contaminants within the aqueous phase in the fracture system was considered. Diffusion of aqueous phase contaminants into the basalt matrix was neglected. As appropriate, radioactive decay and ingrowth also were included.

The transport conceptual model was further expanded to account for contaminants such as volatile organic compounds (VOCs) that migrate in both the dissolved and vapor phase. This expanded model used the dissolved phase transport conceptual model and further included vapor phase advection, dispersion, and diffusion in a dual porosity system. Partitioning of the VOCs between the aqueous and vapor phases was included. Tortuosity of the porous media was used to modify the mobility of the volatile contaminants in the vapor phase. Influences from barometric pressure fluctuations, air injection from drilling of ~40 wells by air-rotary methods, past vapor extraction events, and buoyancy effects also were included. Release of VOCs to the atmosphere was by both advection and diffusion in the vapor phase. The source term release conceptual model, development and implementation for this SDA simulation study is presented in a separate companion paper.

SIMULATION CODE

The TETRAD code (Vinsome and Shook 1993) was chosen to simulate flow and transport. TETRAD has complete multi-phase, multi-component simulation capabilities. Movement of any number of components within aqueous, gaseous, and oleic phases were considered in the SDA simulation study. TETRAD uses a block-centered finite-difference approach and has capabilities for local grid refinement, which were used extensively. The TETRAD simulator also includes dual porosity simulation capabilities. This feature was used to address gaseous-phase movement in both the fracture and matrix portions of the fractured basalts comprising the majority of the subsurface beneath the RWMC.

FLOW MODEL

Stratigraphic information on ~90 wells in the RWMC vicinity from a United States Geological Survey database were used to assign lithologic properties to the three-dimensional simulation domain. A base grid encompassing the region from north of the RWMC to the southern INEEL boundary was defined. Three levels of grid refinement were used in the vicinity of the RWMC to provide increased numerical resolution. This refinement was done both vertically and horizontally. Kriging using detrended lithologic data was used to define the depths of the surficial sediments, and the upper surfaces and thickness of the interbeds: These interpreted surfaces were then used to define material property boundaries in the discretized model (Figure 1).

Hydrologic property estimates came from site-specific analyses. Surface boundary conditions consisted of spatially-varying infiltration based on the results of neutron access-tube monitoring at twenty locations inside the . Three historical flooding events were also included in the simulation. The transient descriptions of infiltration resulted in pulses of water that infiltrated but were damped out with increasing depth as successive interbeds were encountered.

Measured SRPA water levels were used to assign prescribed head boundaries on the sides of the saturated portion of the simulation domain. Three permeability regions were assigned to achieve adequate agreement between simulated and measured aquifer water levels.

Calibration objectives in the vadose zone were the spatial occurrence and temporal behavior of perched water that occurs in association with the interbeds. The perched water behavior in the surficial sediments was not used since it was part of the infiltration model calibration. Although the final calibrated results from the flow model were judged to be adequate for simulating fate and transport, the level of discretization in the deeper interbeds was not adequate to allow detailed modeling of perched water behavior.

DISSOLVED-PHASE TRANSPORT MODEL

Fifty two contaminants were identified for modeling as part of the IRA. From these contaminants, nitrate was selected as the best candidate for calibration of the dissolved-phase transport model. Based on the aquifer monitoring results, there was the possibility of a contribution to nitrate concentrations within the SRPA from the RWMC (Figure 2) that could be used for calibration of the dissolved-phase transport model. This determination was complicated by the presence of nitrate in the aquifer from upgradient (relative to flow within the SRPA) sources. An estimate of the SDA local background concentration of 700 μg-N/L was made by Burgess (1996). This local background concentration was assumed to be correct and the portion of the nitrate concentrations above this level were assumed to come from the SDA for purposes of model calibration. This was a conservative assumption since it implies there is already a contribution from the SDA when it cannot be stated unequivocally that there is such a contribution. There is a markedly small number of samples that have been taken from the vadose zone that could be used for transport calibration for any

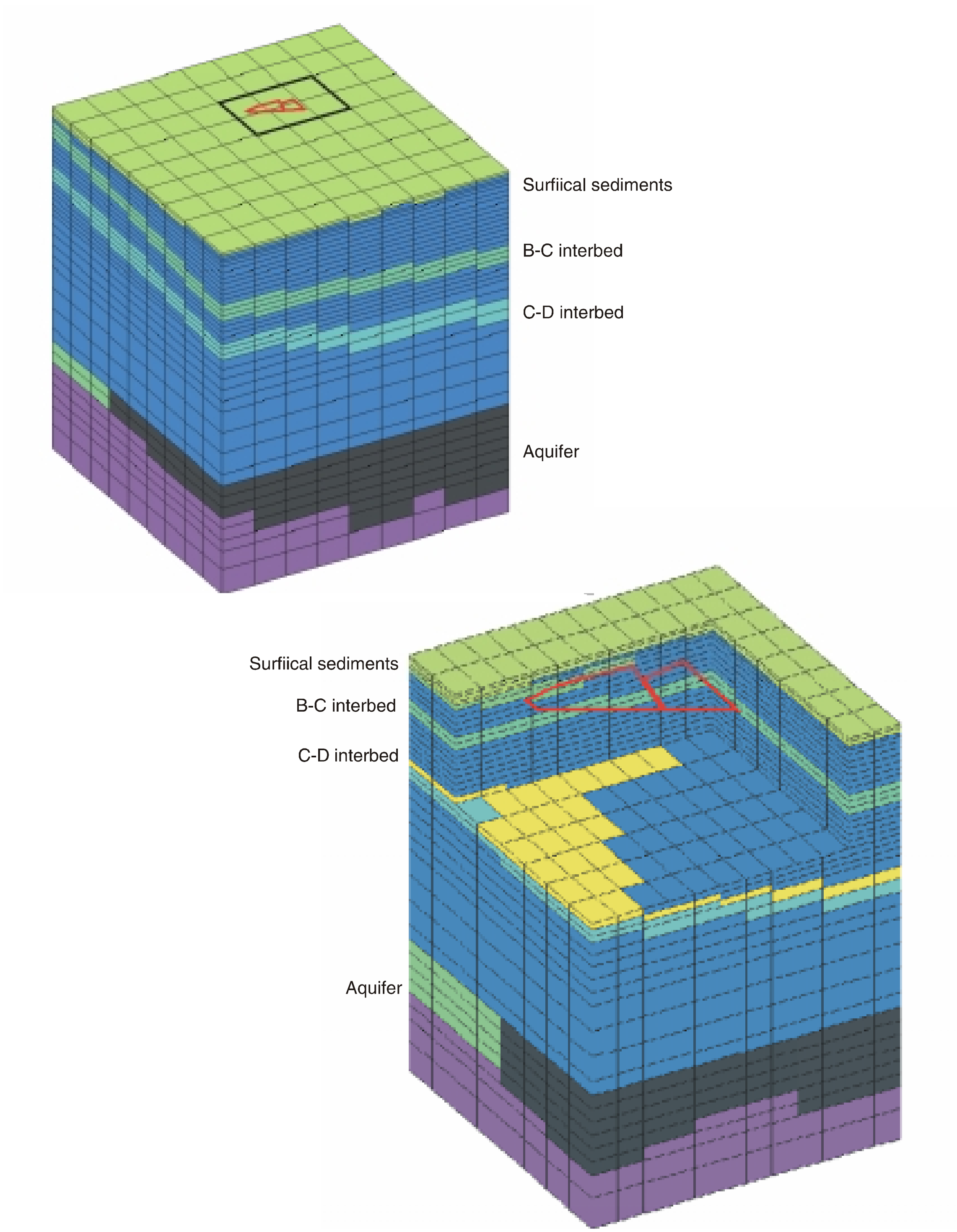

Figure 1. Lithologic assignment in base and first level of grid refinement with cutaway.

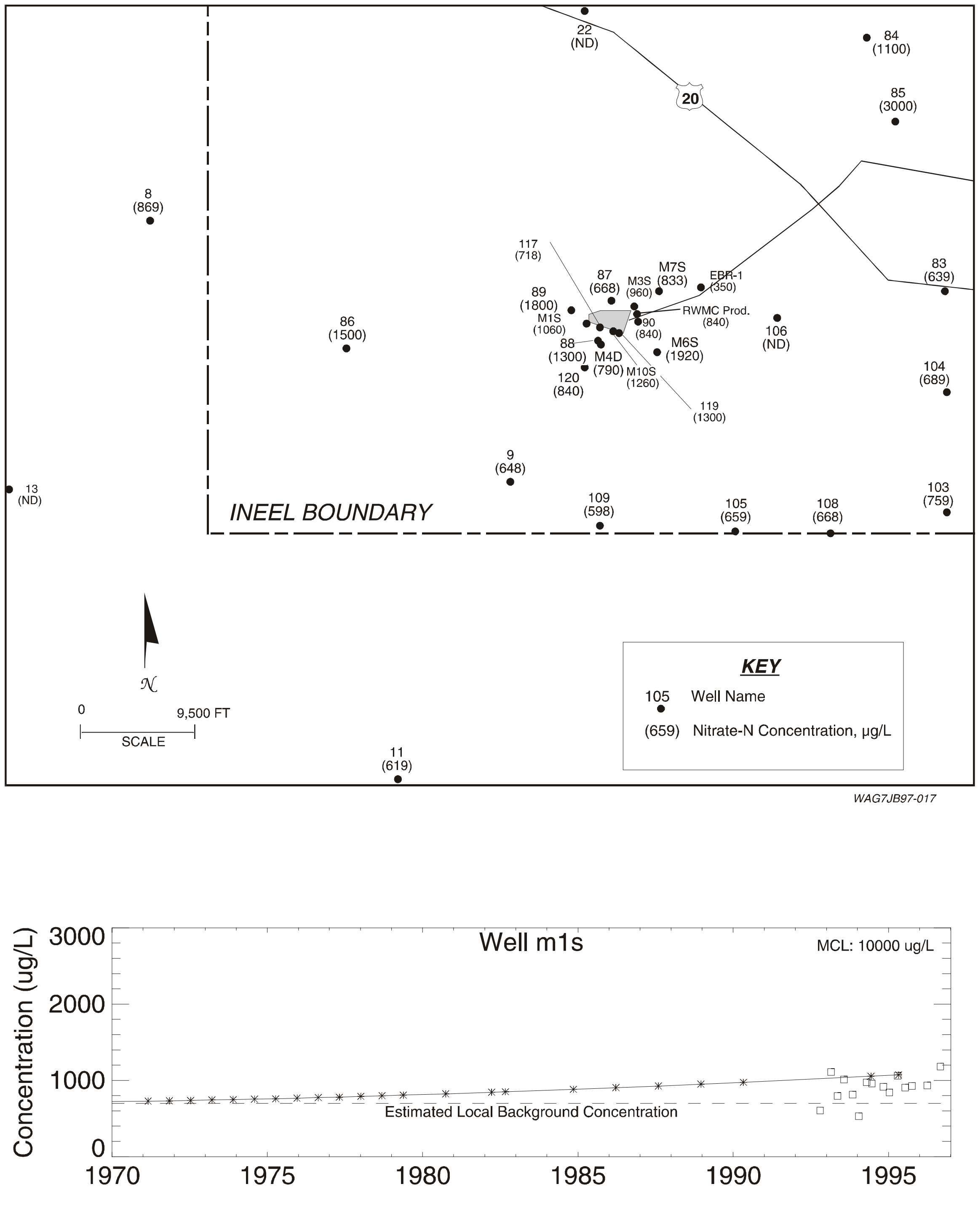

Figure 2. Distribution of nitrate in groundwater in the southwestern INEEL based on sampling in 1994 and 1995 and simulated (asterisks) and measured (boxes) nitrate aquifer concentrations in one well.

contaminant and especially for nitrate since nitric acid has been added as a preservative for perched water samples.

The nitrate release estimated from the source term model was spatially distributed across the SDA (Figure 3). Best-estimates of the nitrate inventory disposed of in the SDA were used for the transport calibration. The calibrated flow model was used and dispersivities were adjusted in the aquifer to improve the agreement between measured and simulated results. Figure 3 also shows a comparison of measured and simulated results for one well as an example.

Primary observations from the dissolved-phase transport modeling were: 1) the model predicted nitrate concentrations to be increasing in the aquifer, and 2) the transport model did a reasonable job of emulating the portion of the nitrate concentrations that were assumed to be from wastes buried in the SDA.

COMBINED AQUEOUS- AND GASEOUS-PHASE TRANSPORT MODEL

The calibrated dissolved-phase transport model was further used to calibrate a transport model for VOCs. VOC source locations (Figure 3) were based on historic disposal data, soil gas survey information, and model calibration results. Carbon tetrachloride (CCl_4) was chosen to calibrate the VOC transport model because the disposal history was relatively well known and it is prevalent in the vadose zone and groundwater. Carbon tetrachloride release was governed by the inventory, drum failure rate, and diffusion from a sludge waste form.

Calibration simulation results were compared to averaged vertical vapor concentration profiles at specific wells, and time histories of vapor concentrations at specific ports, and time histories of groundwater concentrations at specific wells. Figure 4 shows comparisons for two vadose zone vapor monitoring ports and for two aquifer monitoring wells. Comparisons also were made to surface flux and perched water data, but lesser emphasis was given to these comparisons because of the minimal data set.

The final calibrated model showed agreement in magnitude and trend of CCl_4 measurements in the vadose zone and in the aquifer. This agreement was best in the high concentration areas at depth beneath the center of the SDA. Grid resolution was inadequate for making comparisons to near surface vapor concentrations.

SENSITIVITY AND UNCERTAINTY

The sensitivity of predicted aquifer contaminant concentrations to selected hydrologic and transport parameters was qualitatively determined during the flow and transport calibration process, which resulted in the following observations:

- The nitrate calibration is uncertain because of limited data that do not clearly indicate a contribution from the SDA to observed aquifer concentrations. By assuming an SDA contribution above the local estimated background, the model was conservative because the contaminants were modeled to have already reached the aquifer by dissolved-phase transport.

- The modeled aquifer concentrations were relatively insensitive to longitudinal dispersivity in general; however, they were somewhat sensitive to transverse dispersion.

- The predicted aquifer concentrations were generally found to be most sensitive to the inventory and simulated source release mechanisms than any of the hydrologic parameters in the subsurface transport model.

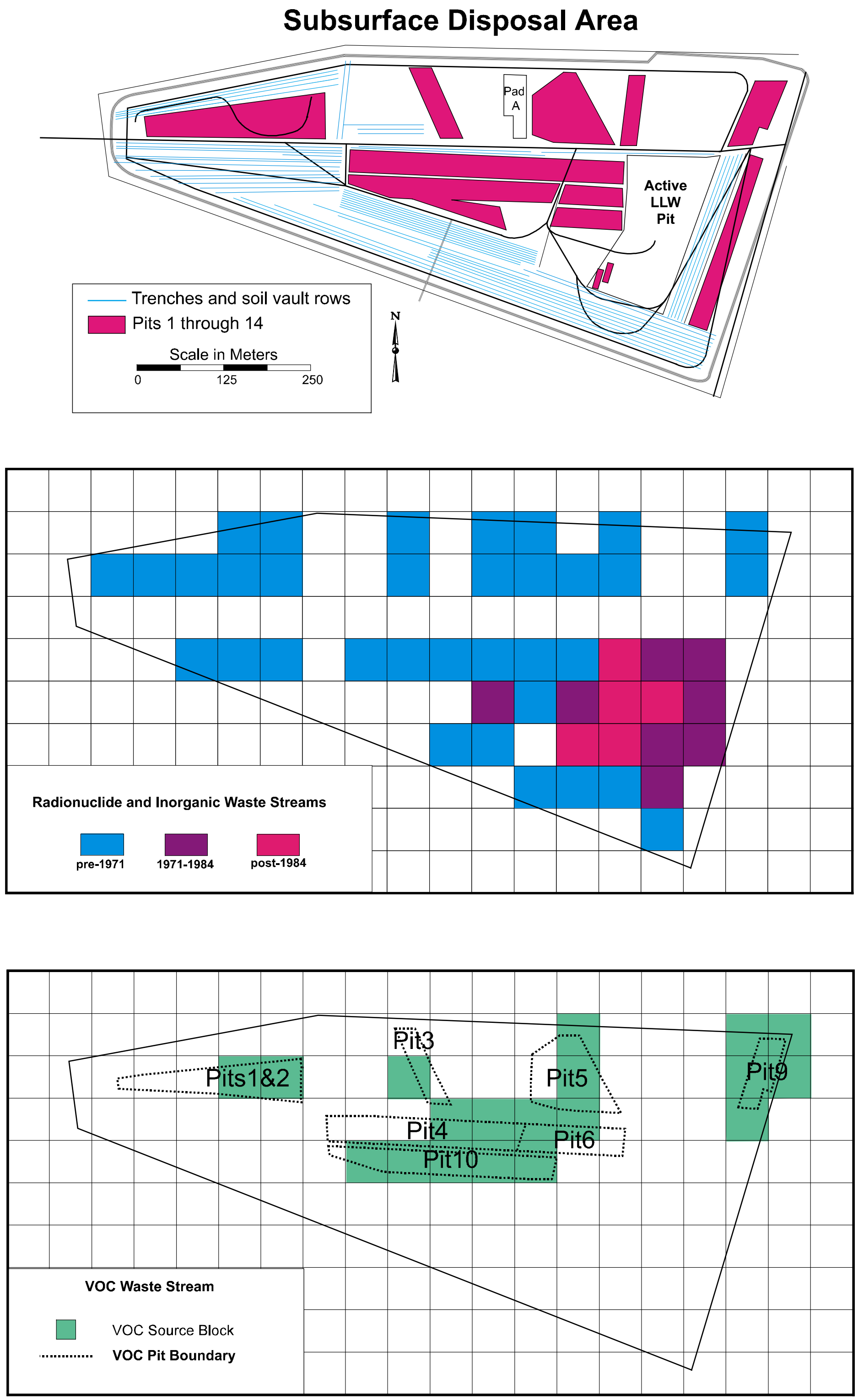

Figure 3. Subsurface disposal area and assignment of contaminant source releases for simulations.

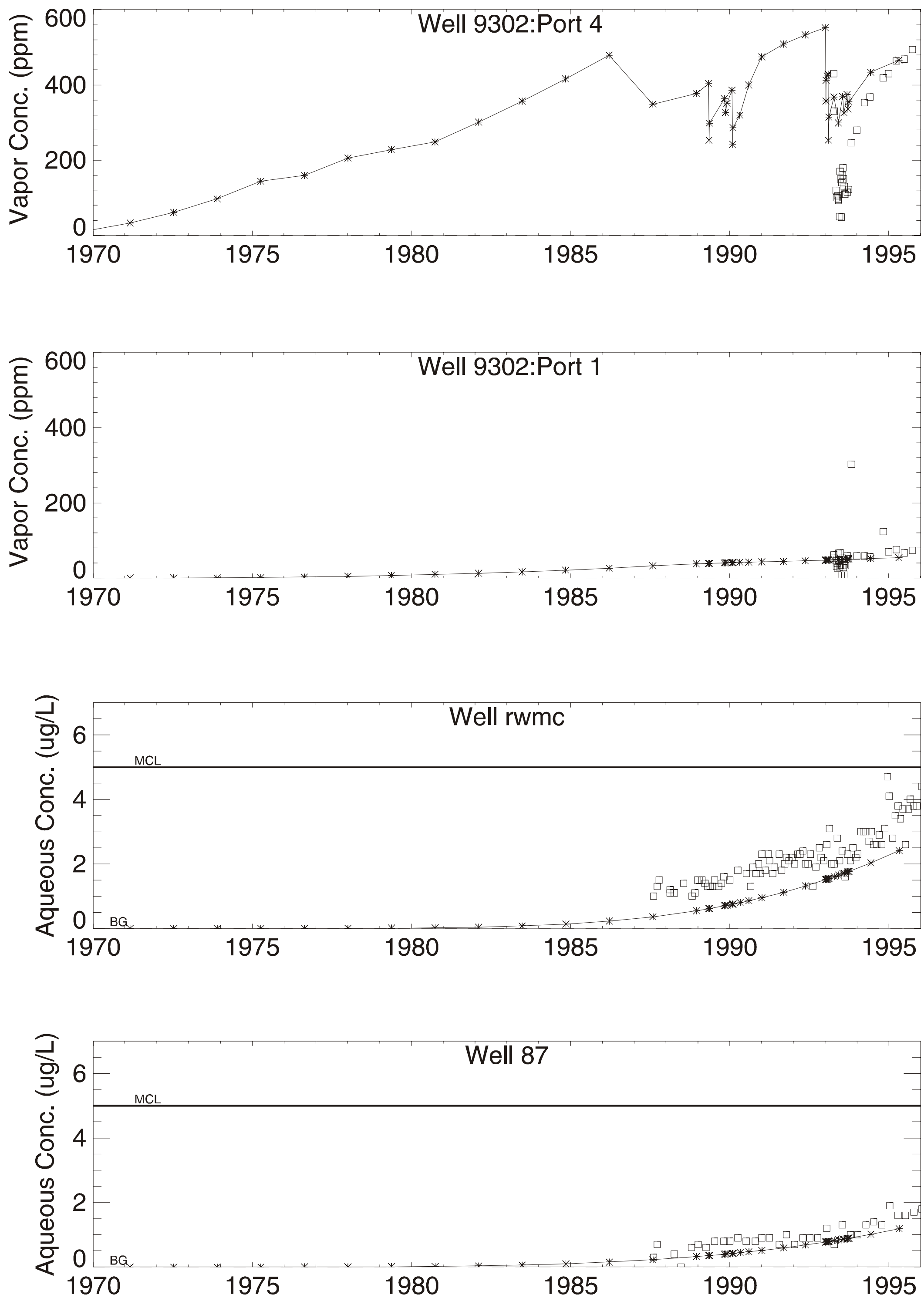

Figure 4. Simulated (asterisks) and measured (boxes) CCl4 vadose zone vapor and aquifer concentrations.

Given the results of the qualitative sensitivity analysis, the largest uncertainties in the predictive simulation results were from uncertainties related to the source term inventory and the description of release mechanisms from buried waste. Uncertainties from the hydraulic and transport parameters used in modeling were smaller than those associated with inventory with the possible exception of some of the partition coefficients that were used.

FACILITATED TRANSPORT STUDIES

A 1996 review of actinide (americium and plutonium) data from subsurface sampling at the Radioactive Waste Management Complex (RWMC) concluded that plutonium had reached the 110-ft and 240-ft interbeds, and that it was unlikely that plutonium detection in samples was a result of downhole or laboratory contamination (3). The review suggested some mechanisms that might explain the transport of plutonium via facilitated transport and recommended additional studies.

Clemson University (CU) and The Florida State University (FSU) have been performing laboratory and computer studies, respectively, for the INEEL since the beginning of FY-97. The purpose of these studies is to determine if high mobility forms of plutonium, such as soluble complexes and/or colloids, are possible transport mechanisms in the subsurface at the INEEL. An external actinide migration peer review panel, consisting of Professor Greg Choppin (FSU), Dr. Jess Cleveland (Consultant), Professor Ken Czerwinski (MIT), and Professor Bob Fjeld (CU), was also organized to review CU, FSU and other studies.

Initial CU column studies, using synthetic perched water and RWMC sedimentary interbed material, suggest the presence of multiple physical/chemical forms having different mobilities depending on the oxidation state of the plutonium and the geohydrological history of the interbed (4). The studies also show, in general agreement with work at other locations in the world, that colloids appear to play an important role in plutonium mobility. The implication of the results, if they are representative of processes that occur under actual field conditions, is that transport models based on a single retardation factor inferred from batch distribution coefficient measurements are not appropriate.

The FSU modeling studies were performed to determine species which are of importance to plutonium transport at the RWMC using the latest thermodynamic values and the USGS computer program PHREEQC (5). Preliminary calculations indicate that plutonium(V) is the most stable oxidation state in RWMC perched water. Complexation by carbonate and fluoride increased Pu(IV) and Pu(VI) concentrations at the expense of Pu(V). Further modeling is underway to gain a better understanding of the role of complexing with selected organic compounds, such as EDTA.

Based on the forgoing, it appears that there are possible physical and/or chemical mechanisms for the transport of plutonium by subsurface waters at the RWMC. However, these mechanisms may or may not be taking place under actual field conditions, or may have only occurred in the past under flooding conditions. There are also other explanations which could account for part or perhaps all of the transport of plutonium to the interbeds, such as through subsurface fractures and/or channels on the outside of well casings.

Further laboratory and computer modeling studies are needed to more fully understand transport mechanisms. It is also equally important to perform extensive sampling under the waste to characterize the actual migration of the plutonium and to determine its speciation in the environment.

CONCLUSIONS

This simulation study represented the first attempt to make an all-inclusive effort to simulate contaminant transport of wastes buried in the SDA. Lack of sufficient data from the vadose zone beneath the SDA limit the ability to completely calibrate a transport model. Although there is considerably more aquifer data available, no clear contribution from the SDA can be discerned because of contamination from upgradient facilities.

The process of performing this SDA simulation study has in part been responsible for the initiation of the following additional or continued characterization activities:

- Sampling will continue in available vadose zone locations utilizing existing suction lysimeters and perched water wells. Sample results in the vadose zone are beneficial for model since they are closer to the source release area and are not subject to interpretation complications from upgradient sources relative to flow in the SRPA.

- Additional upgradient aquifer monitoring wells have been installed to allow determination of possible contribution from SDA waste migration to aquifer concentrations.

- Drilling through waste pits has been proposed to collect samples for determination of contaminant geochemical form and mobility.

- Faciliated transport of a portion of the plutonium isotopes disposed in the SDA has been hypothesized. Studies are underway to assess the likelihood of and contributions from facilitated transport

Results from these additional activities will be useful in either validating the results of this simulation study or guiding future studies.

REFERENCES

1. S.O. MAGNUSON and A. J. SONDRUP, 1998, “Development, Calibration, and Predictive Results of a Simulator for Subsurface Pathway Fate and Transport of Aqueous- and Gaseous-Phase Contaminants in the Subsurface Disposal Area at the Idaho National Engineering and Environmental Laboratory,” INEEL/EXT-97-00609, Lockheed Martin Idaho Technologies Company, July.

2. P.K.W. VINSOME and G.M. SHOOK, 1993, “Multi-Purpose Simulation,” Journal of Petroleum Science and Engineering, Volume 9, pp. 29-38.

3. J. D. Navratil, private communication.

4. R. A. Fjeld, J. T. Coates, A. W. Elzerman and J. D. Navratil, “Column Studies of Plutonium Transport in Sedimentary Interbed From the Snake River Plain,” Proceedings of Waste Management ‘98.

5. M. Bronikowski, G. R. Choppin, and J.D. Navratil, to be published.